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July 14, 2009

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Log Number of Paper: 394

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Further development of Accelerator Mass Spectrometry for the Measurement of Sr-90 at Lawrence Livermore National Laboratory

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Abstract

Based on the encouraging results of our initial efforts to develop a ^{90}Sr accelerator mass spectrometry capability, we have undertaken efforts to enhance our system. By changing some key operating parameters and constructing an optimized detector we were able to improve the discrimination of ^{90}Sr from the isobaric interference ^{90}Zr and reduce our instrumental background by nearly two orders of magnitude. Our current background (4 x 10^6 atoms, 3 mBq) is comparable to that achievable by decay counting, but is still a factor of ten higher than what is theoretically predicted based on the efficiency of our system. Therefore, future plans include implementation of a time-of-flight system to improve the rejection of ^{90}Zr.

Introduction

Strontium-90 ($t_{1/2}$ = 28.78 a) is one of the most important radionuclides managed by agencies charged with protecting the public from radiation such as the United States Department of Energy and the International Atomic Energy Agency. An accelerator mass spectrometry (AMS) method for determination of ^{90}Sr has several potential advantages over conventional radiochemical methods. Detection limits for AMS are typically ~10^5 atoms, equivalent to 0.1 mBq for ^{90}Sr, while detection limits for decay counting are on the order of several mBq[1-3]. Since atoms are counted rather than decay events, AMS offers the possibility to measure low-level samples in minutes rather than the hours or even days required by decay counting[4-5]. Finally, samples can be measured by AMS immediately following sample preparation without waiting for secular equilibrium with ^{90}Y daughter as is the case with decay counting. Based on these potential advantages, we have started developing an AMS method for ^{90}Sr at the Lawrence Livermore National Laboratory's Center for Accelerator Mass Spectrometry.

Our initial development efforts focused on determining optimum system settings (e.g., source target material, molecular ion to inject, terminal voltage/charge state combination, etc.)[6]. Due to the pervasive isobaric interference [90]Zr, our instrumental background for ^{90}Sr was limited to ~10^8 atoms (75 mBq). We have since endeavored to refine our system with the goal of obtaining instrumental backgrounds of at least 10^6 atoms which represents comparable sensitivity to decay counting. This paper will describe our recent system refinement efforts, most notably the implementation of an improved detector design for isobaric discrimination of [90]Zr and [90]Sr.

Experimental

Our experimental setup for the measurement of ^{90}Sr by AMS has been described previously[6], however we have changed some of the key operating parameters in order to improve performance. A brief discussion of the rationale behind these changes follows.

Despite being separated by 2 atomic numbers, high ion energies are required to attain reasonable separation between ^{90}Sr and ^{90}Zr based on differential energy loss in a gas ionization detector. For this reason we have switched the analyzed charge state from 7+ to 11+ which increased the ion beam energy by 57%. Recent refurbishment of our accelerator tubes initially limited the maximum attainable terminal voltage to 8.3 MV. To gain additional ion energy and to compensate for the lower stripping yield associated with the 11+ charge state, we increased the terminal voltage to 9.25 MV which gives a total ion energy of 107 MeV. Eventually, we hope to run our system at 9.75 MV which is the maximum energy of ^{90}Sr^{11+} that can be transported through our high-energy beamline. Finally, we have changed from gas stripping to foil stripping. At the lower terminal voltage and charge state, we were forced to employ gas stripping because Coulomb explosion from foil stripping produced a beam that could not be transported through the high-energy beamline with high transmission.

In addition to the changes discussed above, we invested significant time designing and constructing a new detector to improve the discrimination of ^{90}Sr from ^{90}Zr. In our

initial ^{90}Sr AMS development efforts we employed the same detector that was built for actinide AMS at LLNL and, not surprisingly, this detector was not well suited for ^{90}Sr as demonstrated previously[6]. The new detector, illustrated in Figure 1, incorporates several features intended to optimize the measurement of ^{90}Sr. First, the 1.7-μm thick aluminized Mylar entrance window was replaced with a 200-nm thick silicon nitride window coated with a 10-nm thick layer of aluminum. Silicon nitride has typically been employed to minimize the energy lost in the detector window for AMS at very low energies. Energy loss in the entrance window is not a significant issue in the high-energy regime of AMS, so the primary advantage to the new window is the uniformity and rigidity of silicon nitride[7]. This dramatically reduces the energy straggling in the window and therefore increases the energy resolution of the detector, especially in the first anode.

The second difference between the actinide and ^{90}Sr detectors is the orientation and geometry of the electrodes. The longitudinal-field orientation of the actinide detector was selected for convenience and is adequate for the measurement of actinides where stable isobars do not interfere with the measurement of the isotope of interest. In this case, the pressure inside the detector is set such that the ions of interest lose most of their incident energy in the first anode region and stop within the first few millimeters of the second anode region. Because any interfering ions that enter the detector have vastly different energy or mass than the ions of interest, they are effectively discriminated by requiring coincident signals from both anodes and by drawing an appropriate gate in the energy-loss spectrum.

This approach is ineffective in discriminating ^{90}Sr from the stable isobar ^{90}Zr, so we modified the design of the detector by changing the relative dimensions of the first and second anode to maximize the differential energy loss between ^{90}Sr and ^{90}Zr as illustrated in Figure 2. We also changed the orientation of the electrodes so that the ion path is normal to the electric field lines and we employed a Frisch Grid. These modifications were intended to improve the energy resolution of the detector. Even though this arrangement gives the best separation between the ^{90}Sr and the ^{90}Zr peaks in the energy-loss spectrum, the count-rate from ^{90}Zr is high enough to cause unacceptable dead-times in the signal processing electronics. We therefore added a very short (1.3 cm) third anode positioned just beyond the mean end of range of ^{90}Zr and only process signals that are coincident with all three anodes. This measurement setup effectively reduces the number of processed ^{90}Zr events by ~80% while retaining ~80% of the events from the ^{90}Sr ions of interest.

Results and Discussion

By implementing the changes described above, we were able to significantly improve the discrimination of ^{90}Sr from ^{90}Zr which effectively reduced the instrumental background of our AMS system by nearly two orders of magnitude. Figure 3 shows the two-dimensional energy-loss spectra (Anode 1 vs. Anode 2+3) obtained from a radiochemical standard and a blank measured in our new ^{90}Sr detector. In contrast to the spectra we obtained previously, there are clearly distinct peaks corresponding to ^{90}Sr and ^{90}Zr. However, since the peaks are not completely resolved, the software gate must be

carefully drawn so that as many ^{90}Zr counts are rejected as possible without excluding any more ^{90}Sr counts than necessary. In order to assess the performance of our measurement setup, we measured a series of radiochemical standards and several blanks (Figure 4). The linear fit of the calibration curve is good and the mean value of the blanks corresponds to a ^{90}Sr/Sr of 5.98 x 10^{-13}. Based on the mass of sample loaded into our ion source (equivalent to 1 mg of Sr), this translates into a background of 4 x 10^{6} atoms (3 mBq).

Conclusions

We have made significant improvements to our AMS system for the measurement of ^{90}Sr. By changing key operating parameters and building an optimized detector, we have reduced our instrumental background down to 4 x 10^{6} atoms, which represents an enhancement of nearly two orders of magnitude. This level of sensitivity is comparable to what is achievable by decay counting, however the measurement time for AMS (minutes) is much faster. Despite the improvements we made, our instrumental background is still limited by isobaric interference from ^{90}Zr and is about a factor of ten above the theoretical limit based on the efficiency of our system. Therefore, future plans include addition of a time-of-flight system to our detector in order to add further rejection of ^{90}Zr. We have also begun developing chemical protocols for the preparation of real samples. Our efforts have been based on the rapid solid-phase extraction columns available from Eichrom (Lisle, IL) that were developed for purification of ^{90}Sr for decay counting applications. However, since Zr is not generally an interference for decay

counting, the standard methods for these columns do not appear to have sufficient suppression of ^{90}Zr for AMS. We are therefore investigating adaptations of the standard methods.

Acknowledgements

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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Figure Captions

Figure 1. A schematic diagram of the gas ionization chamber optimized for ^{90}Sr (A1: Anode 1; A2: Anode 2; A3: Anode 3; FG: Frisch Grid; C: Cathode; EW: Entrance Window, 200-nm thick aluminized silicon nitride). The detector typically operates with 165 Torr of P-10 counting gas.

Figure 2. A plot showing the calculated differential energy loss (-dE/dx) as a function of depth for 107-MeV ^{90}Sr and ^{90}Zr ions travelling through P-10 gas, corrected for the energy loss in the 200-nm silicon nitride entrance window. The data were calculated with SRIM-2003.26[8]. The abbreviations used are consistent with those defined in Fig. 1.

Figure 3. Differential energy loss spectra (Anode 1 vs. Anode 2+3) obtained for a) a radiochemical standard (^{90}Sr/Sr = 1.88x10^{-9}), and b) a blank, measured in the new detector optimized for ^{90}Sr. The abbreviations used are consistent with those defined in Fig. 1.

Figure 4. Calibration curve obtained from measurement of a series of radiochemical standards (circles) and blanks (squares) with our improved ^{90}Sr AMS setup. The error bars reflect the larger of counting statistics (internal error) and measurement reproducibility (external error). For the standards, the precision is typically ~1%.
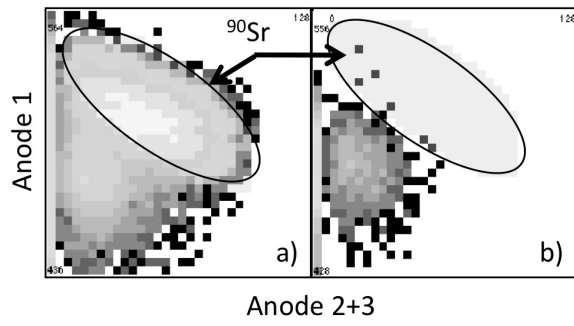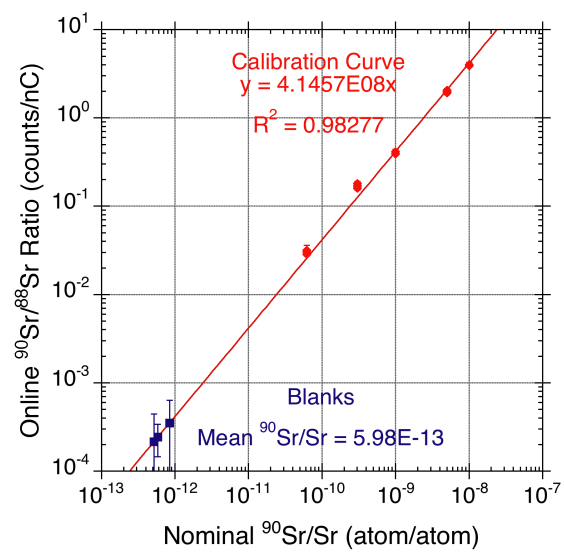
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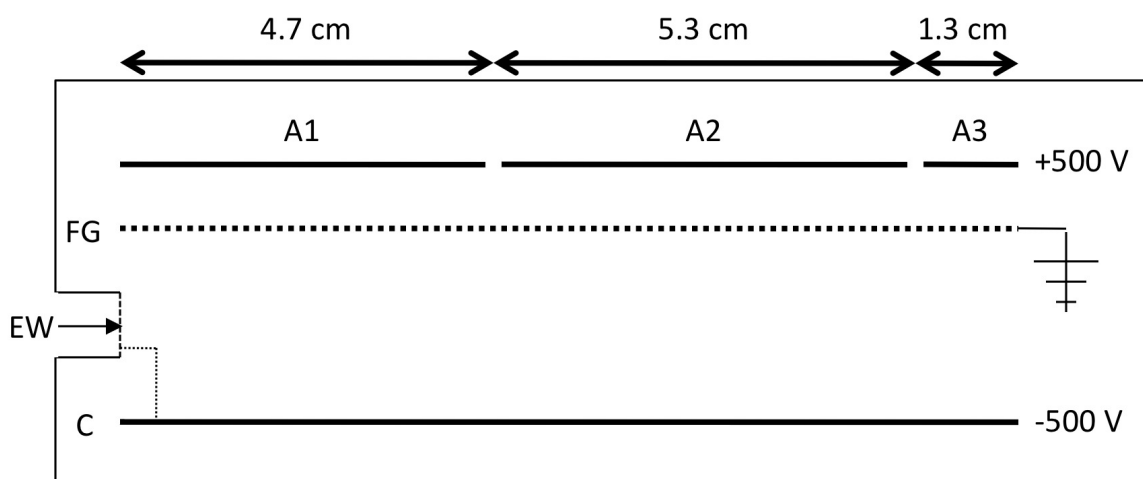
Log 394 – Tumey – Figure 1

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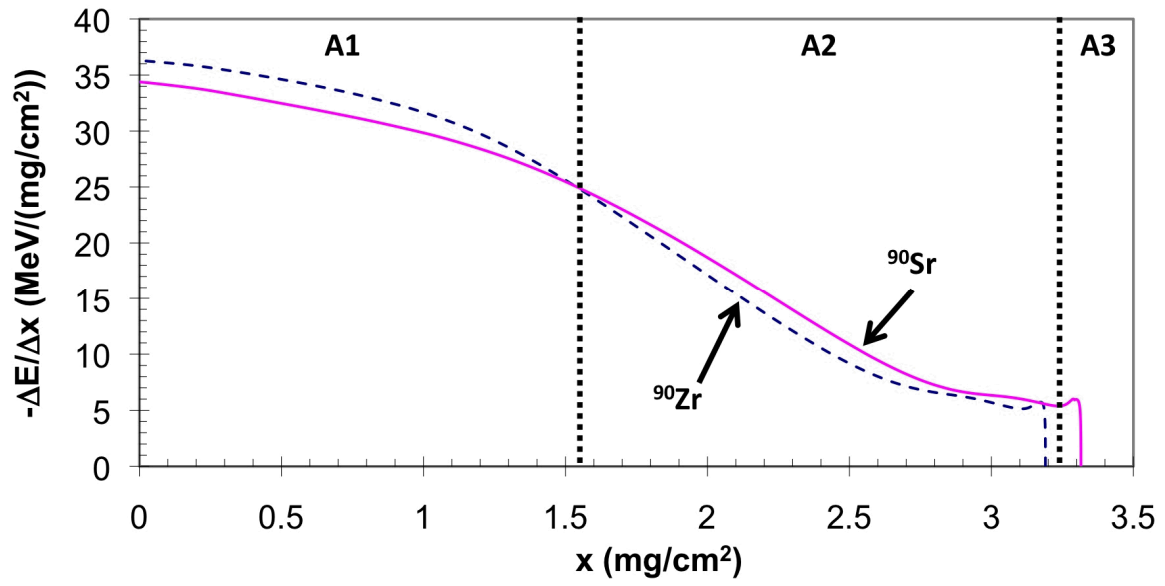
Log 394 – Tumey – Figure 2

Log 394 – Tumey – Figure 3

Log 394 – Tumey – Figure 4